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Relative measurements of tensor components for intrinsic and induced second-order nonlinear susceptibilities in glass optical fibers

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We have measured the x and y components of both the intrinsic and induced second-order nonlinear susceptibilities in glass optical fibers. In addition, we have measured the angular dependence of the intrinsic and induced second-harmonic light intensity as a function of input polarization. Our results are discussed in relation to the models put forward to date to explain second-harmonic generation in optical fibers.

1. Introduction

To date, several models have been put forward [1-5], in an attempt to explain second-harmonic generation (SHG) in glass optical fibers. Due to the complexity of the glass structure most of these models are based upon fairly simplistic assumptions. We have in this paper measured some fundamental properties of the photoinduced SH light, in order to further elucidate the mechanism behind SHG in glass fibers.

The fundamental properties we have chosen to study are the relative magnitudes of the tensor components of the intrinsic and induced second-order nonlinear susceptibilities. The intrinsic SH light is from electric quadrupole interactions at the core-cladding interface of the fiber as predicted by Terhune and Weinberger [6] and recently verified experimentally [7]. The electric quadrupole tensor, which is of rank four, has only three independent

components in glass; due to the constraints imposed by the waveguiding properties of the fiber only the ratio between two of them can be measured in this geometry. Measurements of the induced susceptibility are also limited by the geometry of the waveguide, since the overlap integral is zero for certain polarization combinations.

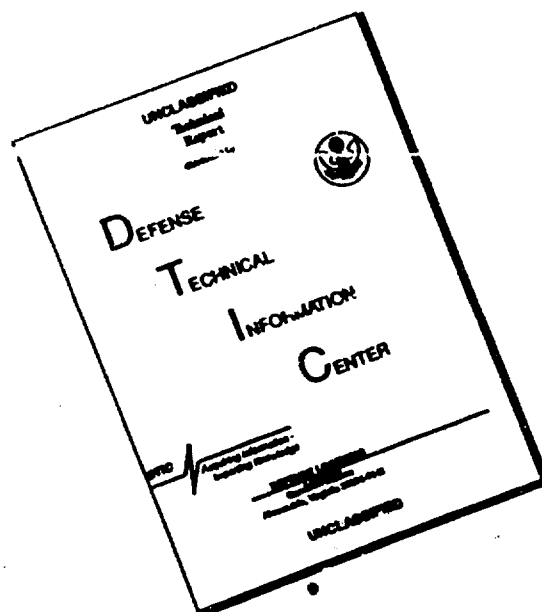
In addition we have measured the angular dependence of the intrinsic and induced SH light intensity as a function of input polarization. The data suggest that the induced SH light comes primarily from the initial electric quadrupole interaction and that growth occurs via improved phasematching. It has already been suggested [8] that an increase in coherence length could explain the increase in the intensity of the SH light. The justification of such a statement is easily obtained from an order of magnitude calculation. In the literature it has been reported that the coherence length can be increased from 30 μm to 30 cm [9] in specially prepared fibers. This in itself would guarantee an increase of the SH light by 8 orders of magnitude - in excellent agreement with experiments.

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2. Theory

One of the problems in measuring a nonlinear susceptibility in a waveguide is the interaction between different modes. Experimentally, one can measure an effective nonlinear susceptibility $\chi_{\text{eff}}^{(2)}$ which consists of the susceptibility times a factor, f , known as the overlap integral. A good review of overlap integrals in waveguides can be found in ref. [10]. For the modes in an optical fiber the effective $\chi_{\text{eff}}^{(2)}$ can be expressed as

$$\chi_{\text{eff}}^{(2)} = \frac{1}{N} \int_{A_x} P_{2\omega} \times H_{2\omega} dx dy, \quad (1)$$

where $dx dy$ is a plane perpendicular to the direction of propagation and N is the normalization factor. The polarization P can be written as

$$P_i = \chi_{ijk}^{(2)} E_j E_k \quad (2)$$

for an electric-dipole interaction and as

$$P_i = \Gamma_{ijkl}^{(2)} E_j \nabla_k E_l \quad (3)$$

for an electric quadrupole interaction. In our calculations we assume the nonlinear susceptibility is constant and therefore move it outside the integral. The remaining integral is the overlap integral denoted f , which we separate into a bulk part and an interface part. The bulk part corresponds to the core of the fiber and the interface part is the region between the core and the cladding estimated from the Goos-Hänchen shift. It is important to note that the interface and bulk susceptibilities have different units related through the thickness of the interface region [11]. In tables 1 and 2 we have listed the calculated

Table 1
Calculated overlap integrals for electric dipole interactions

	x-polarization	y-polarization
ω -He ₁₁ ; ω -He ₁₁ ; 2ω -He ₁₁ core		
$E_x E_x$	3.03	3.03
$E_x E_z$	-6.06×10^{-3}	6.06×10^{-3}
ω -He ₁₁ ; ω -He ₁₁ ; 2ω -He ₂₁ interface		
$E_x E_x$	0.54	0.54
$E_x E_y$	-8.90×10^{-4}	-8.90×10^{-4}
$E_x E_z$	-1.05×10^{-2}	-3.58×10^{-3}

Table 2

Calculated overlap integrals for electric quadrupole interactions

	x-polarization	y-polarization
ω -He ₁₁ ; ω -He ₁₁ ; 2ω -He ₂₁ core		
$E_x E_x$	-4.67	-1.55
$E_x E_z$	-1.50×10^{-3}	-1.50×10^{-3}
$E_z E_z$	9.18×10^5	0
Interface		
$E_x E_x$	-0.24	-8.07×10^{-2}
$E_x E_z$	5.18×10^{-4}	5.18×10^{-4}
$E_z E_z$	2.06×10^5	0

values of the overlap integrals for electric dipole and electric quadrupole interactions separately. We have limited ourselves to LP₀₁ (HE₁₁) and LP₁₁ (HE₂₁, TM₀₁ and TE₀₁) modes since they were the only ones observed in our fibers. The overlap integrals for polarization combinations not listed in tables 1 and 2 are zero. In table 2, although the electric quadrupole interactions are described via a fourth rank tensor (in principle requiring four subindexes) we have limited ourselves to three subindexes in the table for practical reasons; due to the symmetry constraints of the tensor, such a reduction in indexes is unambiguous. Some of these overlap integrals (the ones related to $E(\partial/\partial z)E$ terms) might seem to have unusually large magnitudes. In these cases, the z -derivative multiplies the "geometrical" overlap integral with the propagation constant β .

Another problem of measuring tensor susceptibilities in optical fibers is that in a circular fiber every mode of the light has three directions of polarization (E_x , E_y and E_z). This means that even though our input light is linearly polarized, in the output we inevitably measure a linear combination of tensor components.

There are two ways to simplify the situation: one is to discard all the terms with small values of the overlap integral, and the second is to use a polarization preserving fiber. From the calculations of overlap integrals, it was obvious what terms could be discarded; to implement the second option we used fibers with extinction ratios between the E_x and E_y polarizations that varied between 0.1 and $< 10^{-3}$. In this way we could simultaneously study the influence of the polarization properties of the preparation beam

on the SH light coming out of the fiber. However, it is important to remember that even in the case of a polarization preserving fiber ($E_y/E_x < 10^{-3}$), the ratio E_z/E_x is approximately the same: for our fibers, $E_z < 10^{-5}E_x$. This meant that we could simplify the final expression to read $I_{z\omega}^s = k^2(\Gamma_{xxx}f_{xxx})^2 I_\omega^s I_\omega^s$ and similarly for $I_{z\omega}^s$.

If one attempts to measure an off-diagonal tensor element by exciting the fiber with light polarized 45° to the x -direction it becomes more cumbersome. If we go through the equivalent derivations for this case, it turns out that the only contributing factor to $I_{z\omega}^s$ is the same as for the equation above but with $I_\omega^s = \frac{1}{2}I_\omega^s$. In other words, because of zero-valued overlap integrals, we cannot measure tensor elements such as $\Gamma_{xxy}^{(2)}$ in an optical fiber. To measure $\Gamma_{xxy}^{(2)}$ (and similar off-diagonal tensor elements) we would have to perform our measurements in a bulk glass sample. For electronic dipole interactions we have similar difficulties with overlap integrals being zero. This is why we have omitted the xxx/xyy , etc. ratios in table 4.

It should be pointed out that from these measurements alone, it is in principle impossible to separate one of the components of the forbidden core second harmonic generation from the interface contribution [12]. Our rationale for discussing the core and interface contributions separately are: (i), we have previously given strong evidence that the intrinsic SH light is from a nonlinear interaction at the core-cladding interface [7], and (ii) we have also shown that the magnitude of the intrinsic SH light was proportional to the magnitude of the relative refractive index difference at the interface favoring an electric quadrupole interaction. Also, from a strictly mathematical point of view [12], it is not clear that it is meaningful to separate the interface contributions from dipole and quadrupole interactions as we have done in tables 1 and 2. However, following the analysis of Guyot-Sionnest et al. [13], we believe that separating the interface overlap integrals into dipole and quadrupole interactions gives more physical insight to what is going on at the interface.

3. Experiment

Our measurements were done with a Q -switched and modelocked Nd:YAG laser at 1.2 kHz and 76

MHz respectively. The length of the fibers measured was approximately 7 cm. We used two different types of fiber, one with $V_\omega = 2.49$ and $V_{2\omega} = 5.16$ and a second fiber with $V_\omega = 3.50$ and $V_{2\omega} = 7.20$, where V is the normalized frequency. The input polarization could be arbitrarily varied using a polarizer and a $\lambda/2$ waveplate. For each fiber we characterized the linear polarization properties both before and after "preparation". The polarization extinction ratio varied between 0.1 and $< 10^{-3}$ for the different pieces of fiber used at both the fundamental and SH wavelengths. The preparation was done at many different laser intensities ranging from 0.5 to 20 kW. All fibers were read out at very low fundamental input powers to ensure that no growth took place during the measurements. Although the fibers were always saturated in the x -direction (preparation polarization for all fibers) the read-out was made with the input polarization in a new direction (in which the fiber could be prepared). The various tensor components were measured by setting the input (read-out) polarization to 0° , 45° and 90° , while for each of these input polarizations the output signal was recorded with the analyzer at 0° or 90° . (The x -direction, preparation-direction, corresponds to 0° .) The result of the different intensity ratios from these measurements can be found in table 3 for the intrinsic and photoinduced SH light. In table 4 we have listed the ratios between the equivalent tensor components that we could calculate from tables 1, 2 and 3. The calculations were done using the following formula:

$$\frac{\chi_{ijk}^{(2)}}{\chi_{lmn}^{(2)}} = \frac{f_{lmn}}{f_{ijk}} \left(\frac{I_m^s I_n^s}{I_j^s I_k^s} \right)^{1/2} \left(\frac{I_l^{2\omega}}{I_i^{2\omega}} \right)^{1/2} \quad (4)$$

where f corresponds to an overlap integral and I'' .

Table 3
Measured intensity ratios for intrinsic and indirect SH light after the optical fiber

Ratio of measured intensities	Intrinsic	Induced
I_{xxx}/I_{yxx}	2.4 ± 1.2	30 ± 10
I_{xxx}/I_{xyy}	2.4 ± 1.2	12 ± 3
I_{xxx}/I_{xyx}	2.7 ± 1	3 ± 1
I_{xxx}/I_{yyx}	2.7 ± 1	8 ± 2
I_{xxx}/I_{yyy}	1.5 ± 0.3	155 ± 57

Table 4
Measured ratios between various components of intrinsic and induced second-order nonlinear susceptibilities

Ratio of tensor elements	Intrinsic	Induced
χ_{xxx}/χ_{xx}	1.5 ± 0.4	5.5 ± 1.0
χ_{xxx}/χ_{yy}	1.5 ± 0.4	3.5 ± 0.5
χ_{xxx}/χ_{yy}	1.2 ± 0.3	12 ± 3

$I^{2\omega}$ are the measured light intensities for the fundamental and second-harmonic light, respectively.

We also measured the angular dependence of the intrinsic and induced SH light as a function of input IR polarization as well as output SH polarization. These measurements were performed in the following way. To measure the angular dependency of the SH light as a function of the input polarization we kept the SH analyzer after the fiber in the x -direction ($\theta=0^\circ$) and varied the input IR polarization in increments of 10° . In this measurement $r_{2\omega} = I_{2\omega}(0^\circ)/I_{2\omega}(90^\circ) \approx 10$ for the induced light. The second kind of measurement was to keep the input polarization fixed in the x -direction and vary the SH analyzer after the fiber in increments of 10° , with the result that $r_{2\omega} \approx 30$ for the induced light. The angular dependence for the two measurements is summarized in eq. (5).

$$I(\theta) \approx I_{\min} [1 + r_{2\omega} \cos^4(\theta)] \quad (5)$$

The variation of the intrinsic SH light for different input polarizations also has a $\cos^4(\theta)$ dependence on the input polarization with $r \approx 2.4$. We would like to stress the fact that the ratios given in table 3 were independent of the preparation intensity (the intensity was varied by an order of magnitude) and the degree of polarization preservation (the polarization preservation varied by two orders of magnitude) of the fiber. Another important parameter in our measurements is the number of modes at the fundamental wavelength that propagate through the fiber, since our calculations are contingent upon the fundamental light being in the HE_{11} mode. From measurements of the far-field angle we estimate that less than 10% of the fundamental light could be in a higher order mode. This check should always be made when short pieces of fiber (< 1 m) are being used. The

reason is that a fiber which is nominally "single-mode" will still transmit $\sim 80\%$ of the light excited in the LP_{11} mode if the fiber is, e.g., ~ 10 cm long [14].

4. Discussion

All models put forward to date to explain second-harmonic generation in optical fibers assume the creation of an electric-dipole second-order susceptibility [1-5]. There are primarily two different mechanisms proposed to explain the induced electric-dipole susceptibility. One is that a large ($\sim 10^4$ – 10^5 V/m) electric field is induced into the core of the fiber producing an effective $\chi^{(2)}$ from an allowed $\chi^{(3)}$ interaction (electric-field induced second-harmonic generation). The second mechanism is based on aligning defects in the glass through the intense polarized laser light itself. In view of our findings, we argue against both of these mechanisms.

For the electric-field induced $\chi^{(2)}$, it is reasonable to assume that the dc-field is only in one direction (x for example) which is parallel to the polarization of the light. It that is so, $\chi_{xxx}^{(2)} = \chi_{xyx}^{(2)} = \chi_{yyx}^{(2)} = 0$. This can be deduced from the relationship $\chi_{ijk}^{(2)} = 3\chi_{ijk}^{(3)} E_j E_k E_{dc}$ [15]. Instead (see table 4) we observe that $\chi_{xxx}^{(2)} \sim 0.2\chi_{xxx}^{(2)}$ and $\chi_{yyx} \sim 0.1\chi_{xxx}$. In the dc-field picture, this means that light polarized in the x -direction should generate a dc-field of $\sim 10^4$ V/m in the x -direction and $\sim 10^3$ V/m in the y -direction simultaneously. We find this mechanism highly improbable.

We acknowledge the recent measurements of Mizrahi et al. [16] where they find $\chi_{xxx}^{(2)} \sim 0.008\chi_{xxx}^{(2)}$ in good agreement with a dc-field picture. There is, however, one major difference between these two experiments that we believe can explain the different results.

In the experiment of Mizrahi et al., external seeding at the second-harmonic wavelength was used with a peak power of 20 mW polarized in the x -direction. In our experiment, we used internal seeding which had a peak power between $\sim 10^{-6}$ – 10^{-9} W, depending on which of the two fibers that was used, and, furthermore, this internally generated green light had a third of its power in the y -direction when excited in the x -direction (table 3).

If we now assume that SH in optical fibers occurs through the same physical mechanism whether internal or external seeding is used in the preparation stage, it is clear that the electric field-induced model cannot support both these experimental observations. In the case of external seeding, it seems that using very strongly polarized green light in the preparation stage suppresses the inherent possibility for growth of the perpendicularly polarized green light. If the mechanism of aligning defects were correct, we would again not expect such a large growth of SH light perpendicular to the preparation polarization as we observe.

Based on our measurements, we suggest that the intrinsic electric quadrupole second-order susceptibility is the origin for the photoinduced SH light and that growth occurs through improved phase-matching. We justify this through the remarkably small change in the $\chi_{xxx}^{(2)}/\chi_{xxx}^{(2)}$ and $\chi_{xyy}^{(2)}/\chi_{xxx}^{(2)}$ ratios compared with the overall growth of SH light (7-8 orders of magnitude). We further postulate that the improved phasematching is due to periodic alterations in the electric quadrupole susceptibility along the core-cladding interface so as to cancel the phase-mismatch. In principal, a periodic refractive index could account for the improved phasematching. In this case, the $\Gamma_{\text{eff}}^{(2)}$ would be multiplied by a factor $J_1(\Delta n \kappa A)$ where J_1 is a first-order Bessel function, Δn is the amplitude of the periodic change in refractive index, κ is proportional to $\Gamma^{(2)}$ and A is the periodicity. Typical numbers for an optical fiber give that J_1 is four orders of magnitude too small for this to be feasible. If, instead, the nonlinear susceptibility is modulated, $\Gamma^{(2)}$ will be multiplied by a factor close to one in magnitude. There have been reports of periodic structures in prepared fibers [17], but no experiment has been performed showing what physical parameter is modulated.

In support of our "phase-matching model", we would like to draw attention to the recent discovery that the third-harmonic signal in an optical fiber can also grow with time [18]. To first order this growth was not accompanied with an increase of the third-

order nonlinear susceptibility. This we interpret as another indication that the growth of harmonic signals in optical fibers are primarily from improved phase-matching.

In conclusion, we have measured the ratios of intrinsic and induced nonlinear susceptibilities in two different optical fibers as well as the polarization dependence for the SH light. In addition, we have proposed a new approach in explaining how SH light can grow in an optical fiber.

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